Coherent Backscattering with Nonlinear Atomic Scatterers

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We study coherent backscattering of a quasi-monochromatic laser by a dilute gas of cold two-level atoms. We consider the perturbative regime of weak intensities, where nonlinear effects arise from *inelastic* two-photon scattering processes. Here, coherent backscattering can be formed by interference between *three* different scattering amplitudes. Consequently, if elastically scattered photons are filtered out from the photodetection signal by means of suitable frequency-selective detection, we find the nonlinear backscattering enhancement factor to exceed the linear barrier two.

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Light transport inside a nonlinear medium gives rise to a wide variety of phenomena, like pattern formation, four waves mixing, self focusing effects, dynamical instabilities...[1]. These effects are well described and understood with the help of an intensity dependent susceptibility (e.g. $\chi^{(3)}$ nonlinearity). However, in these approaches, one usually discards the fact that interference phenomena in disordered systems may significantly alter wave transport properties. From this point of view, the most striking systems for which one must combine both nonlinear and disordered descriptions are coherent random lasers [2]. Even if in this case one would require an active (*i.e.* amplifying) medium, the key point is the understanding of the mutual effects between multiple interferences and nonlinear scattering. In particular, in the case of a dilute medium, beyond deviations from the linear regime for the propagation parameters (mean free paths, refraction indices), one may expect modifications of interferential corrections to transport (weak and strong localization regimes). A paradigmatic example is given by *coherent backscattering* (CBS) experiments, where an enhancement of the average intensity scattered around the direction opposite to the incident wave is observed [3]. In the linear scattering regime (where the intensity of the scattered wave is proportional to the incident intensity), CBS arises from interference between *two* reversed scattering paths visiting the same scatterers, but in opposite order. In this case, the CBS enhancement factor, defined as the signal detected in exact backscattering direction divided by the diffuse background, never exceeds two. This maximum value is reached if each pair of interfering paths has the same amplitude, and if single scattering can be suppressed.

Concerning the nonlinear regime, previous studies have been restricted to the case of linear scatterers embedded in a uniform nonlinear medium [4, 5, 6]. Here, it has been shown that the maximum enhancement factor remains two. As we will show in this letter, however, the situation drastically changes in the presence of nonlinear *scattering* (in contrast to nonlinear *propagation*). In particular, in the perturbative regime of at most one scattering event with $\chi^{(3)}$ nonlinearity, CBS arises from interference between *three* amplitudes. Depending on the sign of the nonlinearity, this leads to an increase or decrease of the nonlinear CBS enhancement factor compared to the linear value two. The clearest manifestation of this effect, however, can be observed if the linear component is filtered out from the backscattering signal, such that the latter exclusively originates from nonlinear processes. Then, interference between three amplitudes allows values of the CBS enhancement factor up to three.

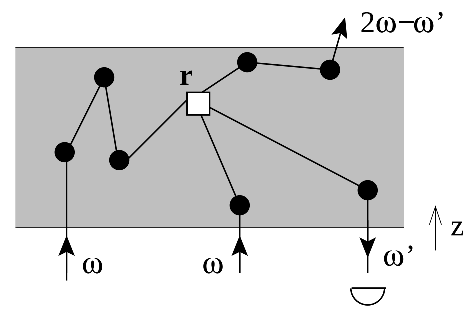

FIG. 1: In the perturbative approach, we assume a single nonlinear two-photon scattering event (□), but arbitrarily many linear scattering events (•). One of the two photons is finally annihilated by the detector, thereby defining the photodetection signal, whereas the other one is scattered into an arbitrary direction.

In the case of a disordered medium consisting of cold two-level atoms, the linear and nonlinear component of the backscattered light can be distinguished in terms of its frequency. This is due to the fact that scattering is purely elastic in the linear regime (described by single-photon scattering), whereas nonlinear multiphoton processes in general change the frequencies of the individual photons, giving rise to inelastic scattering [7]. In this letter, we will restrict ourselves to the perturbative regime of weak laser intensities which is described by scattering of two photons [8]. A typical scattering path is represented in Fig. 1. Here, the two incoming photons propagate at first independently from each other to position

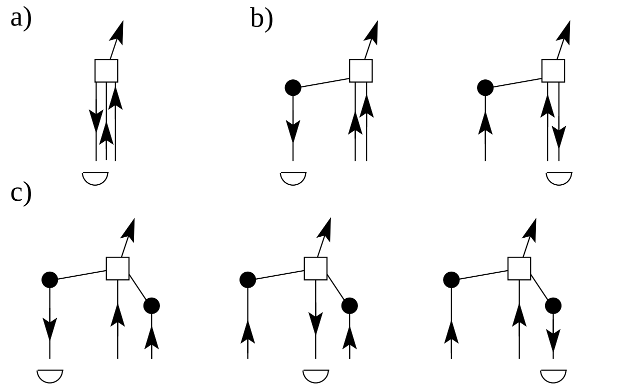

FIG. 2: In the presence of nonlinear scattering (□), there may be either (b) two, or (c) three interfering amplitudes contributing to enhanced backscattering, apart from single scattering (a), which only contributes to the background. In general, the case (c), which corresponds to maximum enhancement factor three, is realized if either both incoming photons, or one incoming and the outgoing detected photon exhibit at least one linear scattering event (•) besides the nonlinear one.

$\mathbf{r}$ inside the disordered atomic medium, where they undergo a nonlinear scattering event. One of the two outgoing photons then propagates back to the detector. The possibility that the two photons meet again at another atom can be neglected in the case of a dilute medium, similar to recurrent scattering in the linear case [9]. We can hence restrict our analysis to processes like the one shown in Fig. 1, with arbitrary number of linear scattering events before and after the nonlinear one. As further discussed below, this perturbative approach is valid for small laser intensity and not too large optical thickness.

To obtain the average intensity measured by the detector placed in backscattering direction, we must identify those scattering paths whose interference survives the ensemble average over the random positions of the scatterers. Again, we consider the case of a dilute medium, where the typical distance between two scattering events (given by the linear mean free path ℓ) is much larger than the wavelength λ. First, we do not obtain interference between diagrams where the nonlinear scattering event occurs at different atoms [8]. Just as in the linear case, we *do* find interference between diagrams where the path of the detected photon is reversed. This gives rise to an enhancement of the average detection signal in exact backscattering direction, on top of the diffuse background. In contrast to the linear case, however, there may be up to *three* different interfering amplitudes, as evident from the examples shown in Fig. 2. In general, the three-amplitudes case is realized if both incoming photons, or one incoming and the outgoing detected photon exhibit at least one linear scattering event besides the nonlinear one.

The perturbative calculation of the backscattering signal relies on the fact that the scattering amplitude of a multiple scattering process like the one shown in Fig. 1 is obtained as a product of the scattering amplitudes for each individual scattering event. The single-photon scattering events can be treated by standard methods known from the theory of multiple scattering in the linear regime [10], which we shortly summarize as follows. Here, the central quantity is the average intensity $I_\omega(\mathbf{r})$ inside the medium. It can be interpreted as the probability that an incoming photon with frequency ω reaches position $\mathbf{r}$ after arbitrarily many linear scattering events, and fulfills the following version of the radiative transfer equation:

$$I_\omega(\mathbf{r}) = e^{-z/\ell} + \mathcal{N}|S_\omega|^2 \int_V d\mathbf{r}'|G_\omega(\mathbf{r},\mathbf{r}')|^2 I_\omega(\mathbf{r}'). \qquad (1)$$

Here, z denotes the distance from the boundary of the medium to $\mathbf{r}$ along the direction of the incident beam, and $\mathcal{N}$ the density of atoms which are randomly distributed in the volume V. Furthermore,

$$S_\omega = \frac{i}{k(1-2i\delta/\Gamma)} \qquad (2)$$

represents the scattering amplitude for a photon with frequency $\omega = kc$ by a single atom, where Γ denotes the width of the atomic resonance at $\omega_{\rm at}$. Eq. (2) is valid in the near-resonant case of small detuning $\delta = \omega - \omega_{\rm at} \ll \omega$. For simplicity, we work with scalar photons, *i.e.*, we neglect the vectorial nature of the light field. In this case, scattering is isotropic, with the total cross section given by $\sigma = 4\pi|S_\omega|^2$. This is not a crucial assumption - the following treatment can be generalized to the vectorial case without serious difficulties. Finally,

$$G_\omega(\mathbf{r},\mathbf{r}') = \frac{e^{in_\omega k|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} \qquad (3)$$

describes average propagation between $\mathbf{r}$ and $\mathbf{r}'$ in the atomic medium, with refractive index given by

$$n_\omega = 1 - \frac{\delta}{\Gamma k\ell} + \frac{i}{2k\ell}. \qquad (4)$$

The imaginary part of n_ω is determined by the linear mean free path $\ell = 1/(\mathcal{N}\sigma)$ at frequency ω.

The first term in Eq. (1) represents the exponential attenuation of the coherent laser mode, *i.e.*, light which has penetrated to position $\mathbf{r}$ without being scattered (Beer-Lambert law). The remaining term describes the diffuse intensity, *i.e.*, light which has been scattered at least once before reaching $\mathbf{r}$. The required solution of Eq. (1) is obtained numerically by iteration, starting from $I_\omega(\mathbf{r}) = 0$. In terms of the average intensity $I_\omega(\mathbf{r})$, the linear components of the backscattering signal are readily obtained by integration over the medium [10].

The second ingredient needed for our nonlinear perturbative analysis is scattering of two photons by a single

atom [8]. Here, the scattered light contains an elastic component, with the same frequency ω as the incident light, and an inelastic component with power spectrum

$$P(\omega') = \frac{\Gamma}{4\pi}\left|\frac{1}{\delta' + i\Gamma/2} + \frac{1}{2\delta - \delta' + i\Gamma/2}\right|^2, \qquad (5)$$

where $\delta' = \omega' - \omega_{\rm at}$ denotes the final detuning. The ratio between the inelastic and elastic component is given by the saturation parameter $s = 2\Omega^2/(4\delta^2 + \Gamma^2)$ (with the Rabi frequency Ω), which measures the intensity of the incident laser in units of the saturation intensity [7].

Combining scattering of two photons by a single atom with the technique for treating the linear case outlined above, we arrive at the following expression for the inelastic background component at frequency ω' (postponing a more detailed derivation to a further publication):

$$L_{\rm in}(\omega') = sP(\omega')\int_V \frac{d\mathbf{r}}{A\ell}\left(2I_\omega^2(\mathbf{r}) - e^{-2z/\ell}\right) I_{\omega'}(\mathbf{r}), \quad (6)$$

where A denotes the transverse area of the medium. The term $2I_\omega^2(\mathbf{r}) - e^{-2z/\ell}$ in Eq. (6) can be interpreted as the average *squared* intensity at position $\mathbf{r}$ inside the slab, which is larger than the square $I_\omega^2(\mathbf{r})$ of the average intensity due to speckle fluctuations [11]. The squared intensity induces a nonlinear atomic response, emitting a photon with frequency distribution $P(\omega')$. Due to time reversal symmetry, the diffusion of this photon from $\mathbf{r}$ to the detector is given by the same expression $I_{\omega'}(\mathbf{r})$ which describes diffusion of incoming photons to $\mathbf{r}$. Note that, in the perturbative regime, the inelastic component contains nonlinear scattering (*i.e.* nonlinearity of the scattering cross section), but not nonlinear average propagation (*i.e.* nonlinearity of the mean free path). Since the latter leaves the photon frequency unchanged, it only affects the elastic component, which we assume to be filtered out from the detection signal.

Concerning the calculation of the interference contribution giving rise to enhanced backscattering, the linear average intensity $I_\omega(\mathbf{r})$, Eq. (1), must be generalized in order to account for different frequencies ω and ω' in the interfering paths:

$$\begin{aligned} g_{\omega,\omega'}(\mathbf{r}) &= e^{i(n_\omega - n^*_{\omega'})kz} + \mathcal{N} S_\omega S^*_{\omega'} \\ &\quad \times \int_V d\mathbf{r}' G_\omega(\mathbf{r},\mathbf{r}') G^*_{\omega'}(\mathbf{r},\mathbf{r}') g_{\omega,\omega'}(\mathbf{r}'). \end{aligned} \quad (7)$$

This function describes the ensemble-averaged product of two probability amplitudes, one representing an incoming photon with frequency ω propagating to position $\mathbf{r}$, and the other one the complex conjugate of a photon with frequency ω' propagating from $\mathbf{r}$ to the detector. If $\omega \neq \omega'$, a nonvanishing phase difference between these amplitudes arises due to both scattering and average propagation in the medium, since both the complex scattering amplitude, Eq. (2), and the refractive index, Eq. (4), depend on frequency. In contrast, the phase difference due to free propagation (*i.e.* in the vacuum) can be neglected if $\Gamma\ell \ll c$, which is fulfilled for typical experimental parameters [12]. In the case $\omega = \omega'$, $g_{\omega,\omega}(\mathbf{r}) = I_\omega(\mathbf{r})$ reduces to the average intensity, see Eq. (1). In terms of the iterative solution of Eq. (7), we find the following expression for the interference term:

$$\begin{aligned} C_{\rm in}(\omega') &= 4sP(\omega')\int_V \frac{d\mathbf{r}}{A\ell}\Big[I_\omega(\mathbf{r})|g_{\omega,\omega'}(\mathbf{r})|^2 \\ &\quad - e^{-z/\ell}{\rm Re}\left\{e^{i(n_\omega - n^*_{\omega'})kz} g^*_{\omega,\omega'}(\mathbf{r})\right\} \\ &\quad - \left(I_\omega(\mathbf{r}) - e^{-z/\ell}\right) e^{-z/\ell - z/\ell'}\Big], \end{aligned} \quad (8)$$

with ℓ' the linear mean free path at frequency ω'. In order to verify that the interference of either three or two amplitudes is correctly taken into account in Eq. (8), it is useful to write the average intensity $I = \exp(-\zeta) + I_D$ as a sum of the coherent mode $\exp(-\zeta)$ (with $\zeta = z/\ell$) plus diffuse intensity I_D. In the case $\omega' = \omega$, we obtain from Eqs. (6,8):

$$\begin{aligned} L_{\rm in} &\propto \left\langle e^{-3\zeta} + 5I_D e^{-2\zeta} + 6I_D^2 e^{-\zeta} + 2I_D^3\right\rangle, \\ C_{\rm in} &\propto \langle \underbrace{\qquad}_{(a)} \underbrace{4I_D e^{-2\zeta}}_{(b)} + \underbrace{12 I_D^2 e^{-\zeta} + 4I_D^3}_{(c)}\rangle, \end{aligned} \quad (9)$$

where the brackets denote the integral over the volume V of the medium, and (a,b,c) correspond to the three cases shown in Fig. 2, identified by different powers of diffuse or coherent light. As expected, the three-amplitudes case (c) implies an interference term twice as large as the background, whereas the case (b) leads to the factor 4/5 known from the scalar two-atom solution (since one of the two interfering amplitudes is twice as large as the other one [8]). Finally, as it should be, the single scattering term is absent in the interference term $C_{\rm in}$.

Using Eqs. (6,8), we now calculate the CBS enhancement factor η for the frequency component ω':

$$\eta(\omega') = 1 + \frac{C_{\rm in}(\omega')}{L_{\rm in}(\omega')}. \qquad (10)$$

The result is shown in Fig. 3, for a slab geometry with three different values of the optical thickness, $b = 0.5$, 1, and 2. Evidently, the largest values of the enhancement factor are obtained if the final frequency ω' approaches the initial one ω, since then the dephasing due to different frequencies vanishes. In this case, the enhancement factor is completely determined by the relative weights between the one-, two- and three-amplitudes cases shown in Fig. 2, cf. Eq. (9). As evident from the dashed line in Fig. 3, already at the rather moderate value $b = 0.5$ of the optical thickness, the three-amplitudes case is sufficiently strong in order to increase the maximum enhancement factor above the linear barrier $\eta = 2$. With increasing optical thickness (and, if necessary, decreasing saturation parameter, in order to stay in the domain

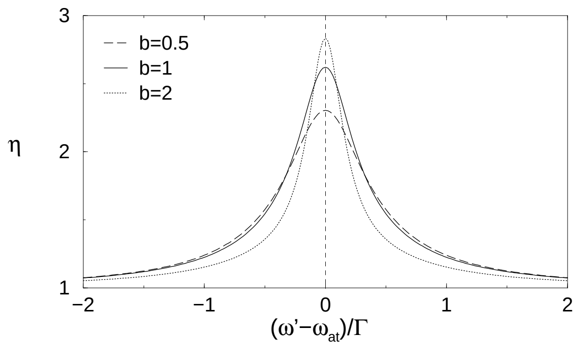

FIG. 3: Spectral dependence of the enhancement factor for coherent backscattering from a slab with optical thickness $b = 0.5$ (dashed line), $b = 1$ (solid line), and $b = 2$ (dotted line) in the perturbative regime of small saturation parameter s. The initial frequency $\omega = \omega_{\rm at}$ is chosen on resonance (*i.e.* $\delta = 0$). The vertical line displays the frequency of elastically scattered photons, which must be filtered out in order to observe an enhancement factor larger than two.

of validity of the perturbative approach, see below), the number of linear scattering events increases, which implies that the three-amplitudes case increasingly dominates, see Fig. 2. In this limit, the enhancement factor approaches the maximum value three. At the same time, however, a larger number of scattering events also leads to stronger dephasing due to different frequencies, $\omega' \neq \omega$. This results in a narrower shape of η as a function of ω' for larger optical thickness. Nevertheless, as evident from Fig. 3, the enhancement factor remains larger than two in a significant range of frequencies ω'. Hence, an experimental demonstration of three-amplitudes interference requires a sufficiently narrow spectral filter, which should be placed as close to the laser frequency ω as possible, but far enough to filter out the elastic component. We have checked that, without filtering, the enhancement factor *decreases* as a function of s (due to the negative sign of the elastic nonlinear component), in qualitative agreement with the experiment [12]. For a quantitative comparison, the elastic component, the geometry of the medium, and the polarization of the photons must be taken into account, which will be presented elsewhere.

Finally, the domain of validity of our perturbative approach, assuming a single nonlinear scattering event, remains to be discussed. A rough quantitative estimation can be given as follows: if p_1 (resp. p_{2+}) denotes the probability for a backscattered photon to undergo one (resp. more than two) nonlinear scattering event, the perturbative condition reads $p_{2+} \ll p_1$. If we assume for all scattering events the same probability s to be nonlinear (thereby neglecting the inhomogeneity of the local intensity), we obtain $p_1 \simeq \langle N\rangle s$, and $p_{2+} \simeq \langle N^2\rangle s^2$, where N denotes the total number of scattering events, and $\langle\ldots\rangle$ the statistical average over all backscattering paths. Evidently, N and N^2 are expected to increase when increasing the optical thickness b. For a slab geometry, we have found numerically that $\langle N\rangle \propto b$ and $\langle N^2\rangle \propto b^3$ (in the limit of large b), concluding that the perturbative treatment is valid if $sb^2 \ll 1$. Let us note that we remain well below the threshold $sb^2 \simeq 1$ at which speckle fluctuations in a nonlinear medium become unstable [13].

A natural way how to extend this work is to give up the perturbative assumption, and admit more than one nonlinear scattering event. This is necessary in order to describe media with large optical thickness, even at small saturation. Since the number of interfering amplitudes is expected to increase if more than two photons are connected by nonlinear scattering events, it may be possible to observe even larger enhancement factors - especially in the case of amplifying scatterers where the nonlinearity appears with positive sign. Finally, the effect demonstrated in this letter might also be relevant for strong localization in the nonlinear regime.

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